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# Simple model of the magnetoelectric effect in layered cylindrical composites

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#### **Abstract**

A simple model of the magnetoelectric (ME) effect in cylindrical layered composites is presented. The cylinder is replaced by the effective layered plate structure with effective length,  $L^{\rm eff}$ , thickness t and height h. In the axial coupling mode, the effective magnetic field acts along the length direction ( $L^{\rm eff}$ ). However, in the vertical coupling mode, the effective magnetic fields act in two directions simultaneously, one along the thickness direction (normal, t) and the other along the effective height (tangential,  $L^{\rm eff}$ ) direction, respectively. Experimental results and theoretical analysis indicated that the cylindrical layered composites ME voltage coefficient is much higher compared with that of the layered plate.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Multiferroic materials exhibit unique magnetoelectric (ME) and mechanical coupling, providing promising opportunities in converting between different energy sources [1]. In the multiferroic materials, the coupling interaction between multiferroic orders could produce some new magnetoelectric or magnetodielectric effects [2]. The ME response is characterized by the appearance of electric polarization upon applying a magnetic field and/or magnetization upon applying an electric field.

Since the 1970s many particulate [3–5] and *in situ* grown [6,7] ME composites have been developed by combining piezoelectric (PE) and magnetostrictive ferrite materials to overcome the limitations of single-phase ME materials, including low ME response and low temperature requirements [8]. However, the ME composites also have some issues with reproducibility and reliability, connectivity control and an insufficient ME voltage coefficient for practical applications [5].

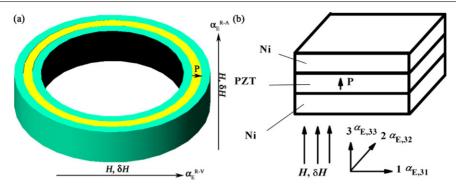
Layered ME composites can overcome problems associated with particulate and *in situ* grown ME compositions. In the past several years, various layered ME composites, such as  $Tb_{1-x}Dy_xFe_{2-y}$  (terfenol-D)/lead zirconate titanate (PZT), ferrite/PZT and terfenol-D/polyvinylidene fluoride (PVDF)

laminates were widely investigated [9–15]. The reported ME voltage coefficient generally levels off at 5.0 V cm<sup>-1</sup> Oe<sup>-1</sup>. Wan et al utilized ME composites bending mode, which largely enhanced the ME voltage coefficient, reaching up to 14.6 V cm<sup>-1</sup> Oe<sup>-1</sup> [16]. One could further increase the ME voltage coefficient by optimizing the configuration of ME composites and utilizing combined loading. Because of the limitations of the preparation method, previous investigations focused only on some simple ME laminate configurations, which included PE/magnetostrictive layers in the form of discs, squares or rectangular shapes. Only recently was cylindrical structure complex configuration studied and its ME behaviour comprehended [17, 18]. In the latest work, we made layered plate and cylindrical ME composites using electro-deposition [17–21]. In this paper, the cylindrical mode is explained by simplifying it as an equivalent plate layered composite bound mode. The mode analysis helps in understanding the cylindrical structure ME properties.

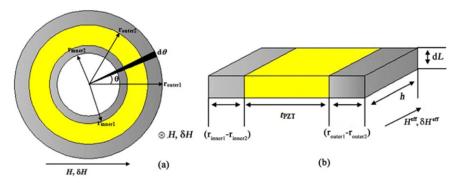
#### 2. Theoretical consideration

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Figure 1 shows cylindrical and plate trilayered ME composite schematics. The cylinder dimensions are  $h \times r_{\text{outer 1}} \times r_{\text{outer 2}} \times r_{\text{inner 1}} \times r_{\text{inner 2}}$ , where h is the cylinder height,  $r_{\text{outer 1}}$ ,  $r_{\text{outer 2}}$ ,  $r_{\text{inner 1}}$  and  $r_{\text{inner 2}}$  are piezomagnetic (PM) layers outer and inner



**Figure 1.** (a) Schematic of the cylindrical layered ME composite.  $\alpha_E = \alpha_E^{R-A}$  and  $\alpha_E = \alpha_E^{R-V}$  when magnetic field is applied in the axial and vertical directions of the cylinder, respectively. (b) Schematic of the plate ME composite structure.  $\alpha_E = \alpha_{E,31}$  and  $\alpha_E = \alpha_{E,32}$  when magnetic field is applied along the length and the width directions of the plane, respectively.



**Figure 2.** (a) Schematic of the differential coefficients method in the axial coupling mode of cylindrical layered composite; (b) schematics of the corresponding simple model for the differential coefficient.

radii, counting from the outside. The middle layer is the PE phase, such as PZT, barium titanate (BT) or other, polarized along the radial direction. Outer and inner layers are made of the PM phase, such as Fe, Co, Ni or an alloy of these elements.

When measuring the ME voltage coefficient of the cylindrical trilayered ME composites, one can apply the bias magnetic field H and the sinusoidal magnetic field SH along two different directions, i.e. the axial coupling mode where H and SH are applied along the cylinder axis, and the vertical coupling mode where SH and SH are applied perpendicular to the cylinder axis. We denote the ME voltage coefficient as  $\alpha_E^{R-A}$  in the axial coupling mode and  $\alpha_E^{R-V}$  in the vertical coupling mode, respectively, and the superscript SH identifies the PE polarization direction (figure SH). The magnetoelectric measurement system has been described in detail elsewhere [22].

In the axial coupling mode, one can deal with the cylindrical trilayered composite using the method of differential coefficients along the toroidal cylinder direction (figure 2(a)) and simplify it as a differential plate trilayered ME composite of  $h \times dL \times t$  dimensions (figure 2(b)).

Planar dimensions in the polar and the Cartesian coordinate systems are related as

$$(r_{\text{outer }1}^2 - r_{\text{inner }2}^2) d\theta = 2(r_{\text{outer }1} - r_{\text{inner }2}) dL,$$
 (1)

where  $d\theta$  is the differential coefficient radian. Since the cylinder is a continuous medium, one can express its effective

length,  $L^{\rm eff}$ , as

$$L^{\text{eff}} = \int_0^{2\pi} dL = \pi (t_{\text{PM}} + t_{\text{PE}} + 2r_{\text{inner 2}}), \tag{2}$$

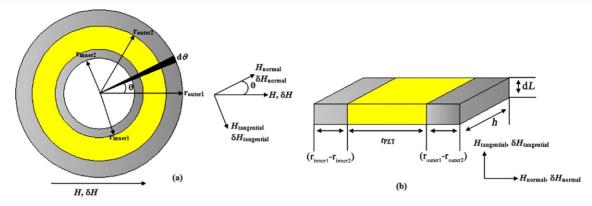
where  $L^{\text{eff}}$  and dL are the effective length and the differential coefficient length of the simplified plate trilayered ME composite, respectively,  $t_{\text{PM}}$  is the total thickness of the PM phase and  $t_{\text{PE}}$  is the thickness of the PE phase. For both the cylinder and the plane,  $t_{\text{PM}} = [(r_{\text{outer 1}} - r_{\text{outer 2}}) + (r_{\text{inner 1}} - r_{\text{inner 2}})]$  and  $t_{\text{PE}} = (r_{\text{outer 2}} - r_{\text{outer 1}})$ .

We denote the effective sinusoidal magnetic field as  $\delta H^{\rm eff}$  and the effective bias magnetic field as  $H^{\rm eff}$  in the simplified mode. Since H and  $\delta H$ , as well as  $\delta H^{\rm eff}$  and  $H^{\rm eff}$ , are perpendicular to the differential coefficient face, i.e. along the h direction, they can be expressed as

$$\delta H^{\text{eff}} = \delta H,$$

$$H^{\text{eff}} = H.$$
(3)

Using this analysis, the cylindrical trilayered ME composite axial coupling mode can be simplified as a plate trilayered ME composite longitudinal mode. The dimensions of the simplified plate trilayered ME composite are  $h \times L^{\rm eff} \times t$ , where t is the total thickness of the PE phase  $t_{\rm PE}$  and the PM phase  $t_{\rm PM}$ . After simplification, the effective magnetic fields  $H^{\rm eff}$  and  $\delta H^{\rm eff}$  are along the h direction, and their values are the same as the measured and the applied magnetic fields.



**Figure 3.** (a) Schematic of the differential coefficient method in vertical coupling mode of cylindrical layered composite; (b) schematic of the corresponding simple model for the differential coefficient.

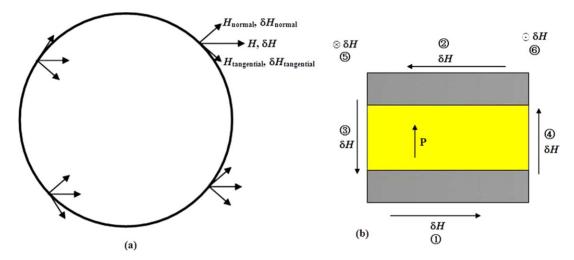


Figure 4. (a) Magnetic fields H and  $\delta H$  that produce normal ( $H_{\text{normal}}$  and  $\delta H_{\text{normal}}$ ) and tangential ( $H_{\text{tangential}}$ ) magnetic fields in different parts of the cylinder; (b) Magnetic fields H and  $\delta H$  directions in a plate trilayered composite.

In the vertical coupling mode, one can also deal with the cylinder using the method of differential coefficients, similar to the axial coupling mode. The cylinder can also be simplified as a differential trilayered plane. However, magnetic fields H and  $\delta H$  are neither perpendicular nor parallel to the layers during measurements. Here, the magnetic fields H and  $\delta H$  can be considered acting along the cylinder normal and tangential directions (figure 3(a)). We denote the normal magnetic fields as  $H_{\text{normal}}$  and  $\delta H_{\text{normal}}$ , and the tangential fields as  $H_{\text{tangential}}$ and  $\delta H_{\text{tangential}}$ . However, for different parts of the cylinder,  $H_{\text{normal}}$  and  $\delta H_{\text{normal}}$  are not equivalent, and the same applies to  $H_{\text{tangential}}$  and  $\delta H_{\text{tangential}}$  (figure 4(a)). At any given time, the opposite sides of the layered ME composite are subjected to magnetic fields H and  $\delta H$  that are equal in magnitude but opposite in direction (figure 4(a)). The ME voltage coefficients are the same for the equal and opposite pairs of fields denoted by encircled numbers in figure 4(b), where 1 and 2 represent the opposite fields along the length of the plane, 3 and 4 along the thickness of the plane and 5 and 6 along the width of the plane. Therefore, we can also consider H and  $\delta H$  in two directions for the simplified mode, i.e.  $H_{normal}$  and  $\delta H_{normal}$  along the thickness direction and  $H_{\text{tangential}}$  and  $\delta H_{\text{tangential}}$  along the differential coefficient length (figure 3(b)). Then one can get

the expressions for the corresponding applied magnetic fields:

$$\delta H_{\text{normal}} = \delta H \cdot |\cos \theta|,$$

$$H_{\text{normal}} = H \cdot |\cos \theta|,$$

$$\delta H_{\text{tangential}} = \delta H \cdot |\sin \theta|,$$

$$H_{\text{tangential}} = H \cdot |\sin \theta|.$$
(4)

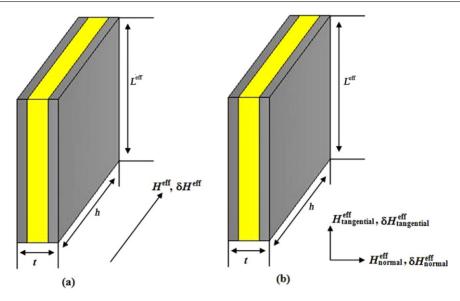
The corresponding effective magnetic fields can be expressed as

$$\int_{0}^{2\pi} \delta H_{\text{normal}} d\theta = 2\pi \cdot \delta H_{\text{normal}}^{\text{eff}},$$

$$\int_{0}^{2\pi} H_{\text{normal}} d\theta = 2\pi \cdot H_{\text{normal}}^{\text{eff}},$$

$$\int_{0}^{2\pi} \delta H_{\text{tangential}} d\theta = 2\pi \cdot \delta H_{\text{tangential}}^{\text{eff}},$$

$$\int_{0}^{2\pi} H_{\text{tangential}} d\theta = 2\pi \cdot H_{\text{tangential}}^{\text{eff}}.$$
(5)



**Figure 5.** (a) Trilayered plate composite simple bound state applied for the cylindrical layered composite in the axial coupling mode; and (b) in the vertical coupling mode.

Combining equations (4) and (5):

$$\delta H_{\text{normal}}^{\text{eff}} = \delta H_{\text{tangential}}^{\text{eff}} = \frac{2}{\pi} \delta H,$$

$$H_{\text{normal}}^{\text{eff}} = H_{\text{tangential}}^{\text{eff}} = \frac{2}{\pi} H.$$
(6)

Similar to the axial coupling mode, one can also get the effective length in the vertical coupling mode. The expression and the values are the same as in the axial coupling mode.

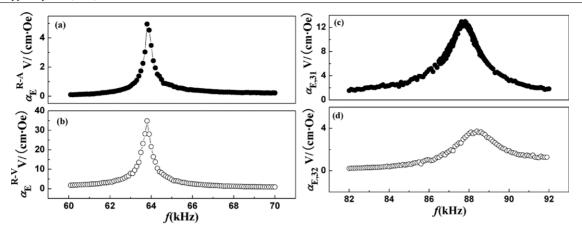
From the above analysis, similarly to the vertical coupling mode, one can also simplify the cylinder as a plate trilayered ME composite with  $h \times L^{\rm eff} \times t$  dimensions. The effective magnetic fields are along the thickness (t) and the effective length  $(L^{\rm eff})$  directions of the plate, simultaneously, and their values are  $2/\pi$  times the applied and measured magnetic fields (figure 5(b)).

#### 3. Results and discussion

In previous work, we studied the ME voltage coefficient of the trilayered cylindrical ME composites in axial and vertical coupling modes. The maximum of  $\alpha_{\rm E}^{\rm R-V}$  was 5 times higher than that of  $\alpha_{\rm E}^{\rm R-A}$  [18] (figures 6(a) and (b)). Lacking an understanding of the cylindrical structure mode, we only reported, but did not explain the result. In this paper, we use the simplified model to explain this phenomenon. Here, we assume that the inner Ni layer thickness is equal to the outer one. Therefore, the sample dimensions can be denoted as  $h \times r_{\text{outer 1}} \times r_{\text{outer 2}} \times r_{\text{inner 1}} \times r_{\text{inner 2}} = 8 \times 10.5 \times 10.5$  $10 \times 9 \times 8.5 \,\mathrm{mm}^3$ . The cylinder can be simplified as a trilayered plate ME composite with dimensions of  $h \times L^{\text{eff}} \times$  $t = 8 \times 59.6 \times 2 \,\mathrm{mm}^3$ . In previous work, we studied the shape demagnetization effect on the ME voltage coefficient of layered ME composites [19]. The ME voltage coefficient strongly depends on the in-plane sample size. The ME effects of the two perpendicular in-plane directions are different if the plate in-plane dimensions are not the same. The paper reported that the peak value of  $\alpha_{E,31}$  is  $14.6\,\mathrm{V\,cm^{-1}\,Oe^{-1}}$ , which is 3 times larger than  $\alpha_{E,32}$  of  $4\,\mathrm{V\,cm^{-1}\,Oe^{-1}}$  for a  $10\times20\times1.6\,\mathrm{mm^3}$  Ni/PZT/Ni trilayered plate ME composite as shown in figures 6(c) and (d). The schematic of the plate ME composite structure is shown in figure 1(b). The results obtained indicate that  $\alpha_{E,31}$  is always larger than  $\alpha_{E,32}$  in the same bias magnetic field. Moreover, previous work indicated that  $\alpha_{E,31}$  was almost one order of magnitude larger than  $\alpha_{E,33}$  for the plate ME composites [20, 23]. Since the cylinder can be simplified as a  $8\times59.6\times2\,\mathrm{mm^3}$  trilayered plate ME composite, and the ME effect of  $\alpha_E^{R-A}$  can be equal to  $\alpha_{E,32}$  and  $\alpha_E^{R-V}$  can be equal to  $\alpha_{E,31}$  and  $\alpha_{E,33}$ , it is not difficult to understand why the maximum of  $\alpha_E^{R-V}$  was higher than that of  $\alpha_E^{R-A}$  in the cylinder.

Although the cylinder can be simplified as an infinitesimally plate layered structure, the boundary conditions of the cylinder are not the same as those of the plate layered structure. Both outer PM layers of the plate structure are under plane stress conditions and are not constrained in the vertical direction (the free vertical state), while the cylinder outer and inner faces are constrained in the axial, radial and circumferential directions (the self-bound state). When the PM ring shrinks (or expands) in the magnetic fields, not only does its circumference decrease (or increase) but also its diameter and height decrease (or increase) at the same time due to the self-bound effect. Then each PE infinitesimal unit will suffer radial and tangential forces simultaneously due to the change in the shape of the PM layers. Two PE modes of  $d_{33}$  and  $d_{31}$ contribute to the ME coefficient at the same time. Hence, the cylindrical layered ME composite can be simplified as a plate trilayered ME composite in the self-bound state [18].

The self-bound state can promote the ME effect in layered composites, which strongly depends on the mechanical coupling between the layers. Guo *et al* reported that the clamped ME composites had a larger ME voltage coefficient than those in the free state [24]. The cylindrical shape forces



**Figure 6.** (a), (b) Frequency dependence of  $\alpha_{\rm E}^{\rm R-A}$  and  $\alpha_{\rm E}^{\rm R-V}$  for the cylindrical trilayered ME composite and (c), (d) [21]  $\alpha_{\rm E,31}$  and  $\alpha_{\rm E,32}$  for the plate trilayered ME composite around electromechanical resonance (EMR) frequency [18].

its infinitesimal elements in the self-bound state naturally, therefore, we predicted that the cylinder would have a much larger ME voltage coefficient than the plate. The maximum of  $\alpha_{E,31}$  was about  $13~V~cm^{-1}~Oe^{-1}$  for the plate sample, while the maximum of  $\alpha_E^{R-V}$  is about  $30~V~cm^{-1}~Oe^{-1}$  for the cylindrical sample with the same magnetostrictive–PE phases thickness ratio [18, 21] (figure 6). Our experimental results are in good agreement with the theoretical predictions.

From the above analysis, the cylinder can be simplified as a plate layered ME composite, whose effective length is larger than the cylinder diameter, leading to enhanced ME properties. Therefore, the cylindrical shape has outstanding merit for miniaturizing ME devices for practical applications. Also, the cylinder is in the self-bound state, where many PE modes contribute to the ME properties at the same time, inducing the ME effect enhancement. For these two reasons, the cylinder has a larger ME voltage coefficient than the plate with the same magnetostrictive—PE phases thickness ratio.

## 4. Summary

In summary, we identified the cylindrical layered mode and simplified it with a self-bound trilayered plate mode with dimensions of  $h \times L^{\rm eff} \times t$  using the method of differential coefficients along the toroidal direction in the cylinder. In the axial coupling mode, the effective magnetic fields were along the length direction. However, in the vertical coupling mode, the effective magnetic fields are acting in two directions simultaneously, along the thickness (normal, t) and the effective length (tangential,  $L^{\rm eff}$ ) direction, respectively. The simplified model of cylindrical layered ME composites is helpful in understanding and enhancing the ME properties and designing appropriate ME devices.

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